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National Dioxin Study Tier 4
Combustion Sources
Project Summary Report

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Research Triangle Park, NC

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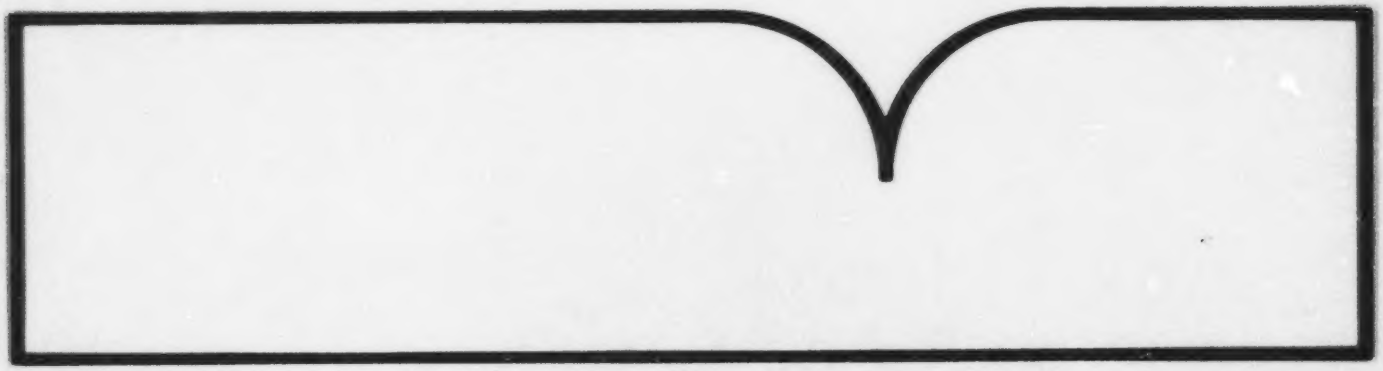

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National Dioxin Study Tier 4 — Combustion Sources

Project Summary Report

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16. ABSTRACT

This report presents the Agency's finding on the assessment of dioxin emissions from combustion sources under Tier 4 of the National Dioxin Strategy. The primary objectives of Tier 4 are outlined in the report. The report describes the information the Agency collected at the request of Congress to improve the current understanding of combustion sources which emit dioxin to the ambient air. The tasks used to collect the information are outlined, including stack testing, ash sampling and data analysis. References to other documents which describe methods, procedures and detailed analyses are identified.

The report finds that small amounts of chlorinated dibenzo-p-dioxins and dibenzofurans are emitted from many combustion sources, with only a few sources emitting larger amounts. These sources include municipal waste incinerators, secondary copper smelters and sewage sludge incinerators.

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National Dioxin Study
Tier 4 — Combustion Sources

Project Summary Report

By
Air Management Technology Branch

U.S. ENVIRONMENTAL PROTECTION AGENCY
Office Of Air And Radiation
Office Of Air Quality Planning And Standards
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The field work was supported by two Office of Research and Development (ORD) Laboratories: (1) the Air and Energy Engineering Research Laboratory, Research Triangle Park, NC, which provided sampling methods support and consultation; and (2) the Hazardous Waste Engineering Research Laboratory in Cincinnati, OH, which provided technical and contractual support for the field testing program. Field support for the collection of samples was also provided by EPA Regional Offices, many State and local environmental agencies, Radian Corporation, and various other supporting contractors.

Analytical support was provided by a group of EPA laboratories, collectively referred to as the Troika, comprising ORD's Environmental Monitoring Systems Laboratory, Research Triangle Park, NC; Environmental Research Laboratory, Duluth, MN; and the Office of Pesticides and Toxic Substance's Environmental Chemistry Laboratory, Bay St. Louis, MS.

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Introduction

This report presents a concise summary of Tier 4, combustion sources, of the U. S. Environmental Protection Agency's National Dioxin Study. It is intended to be an overview document which presents in summary form the major results and conclusions from this study. The major portion of this report is comprised of the chapter on combustion sources taken from the U.S. Environmental Protection Agency's National Dioxin Study Report To Congress presented to the Congress in September 1987.

Objectives

In December 1983, the U. S. Environmental Protection Agency (EPA) issued its National Dioxin Strategy,[1] which was designed (a) to determine the overall extent of dioxin contamination in the environment and (b) to provide a systematic approach for dealing with dioxin contamination problems. The primary focus of the strategy was on 2,3,7,8-tetrachlorodibenzo-p-dioxin (2378-TCDD), which is believed to be the most toxic of the chlorinated dibenzo-p-dioxin (CDD) compounds.* To implement the strategy, the EPA defined the following seven categories (or tiers) of sites for investigation:

Tier 1 - 2,4,5-trichlorophenol (2,4,5-TCP) production sites and associated waste disposal sites. [2378-TCDD is a known contaminant of 2,4,5 trichlorophenols.]

Tier 2 - Sites and associated waste disposal sites where 2,4,5-TCP was used as a precursor to make pesticidal products.

Tier 3 - Sites and associated waste disposal sites where 2,4,5-TCP and its derivatives were formulated into pesticidal products.

Tier 4 - Combustion sources.

Tier 5 - Sites where 2,4,5-TCP and pesticides derived from 2,4,5-TCP have been, or are being, used on a commercial basis.

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*Throughout this report, the abbreviations CDD and CDF are used to indicate chlorinated dibenzo-p-dioxin compounds and chlorinated dibenzofuran compounds, respectively. CDFs are compounds similar to CDDs in structure and chemical activity.

Tier 6 - Sites where improper quality control on manufacturing of certain organic chemicals and pesticides could have resulted in the inadvertent formation of 2378-TCDD.

Tier 7 - Control sites where contamination from 2378-TCDD was not suspected.

This report summarizes the finding of Tier 4, the portion of the study dealing with combustion sources. The primary objective of the Tier 4 study was to determine the potential scope and magnitude of CDD and CDF releases from combustion sources. The study was designed to determine which combustion source categories emit CDDs and at what concentrations. The main focus was on releases to the ambient air; however, other samples, such as ash and scrubber water, were also obtained to determine if these compounds are released to other media. Because some combustion sources were known to emit a wide range of CDD and CDF compounds, Tier 4 samples were analyzed for specific groups (homologues) of CDD and CDF compounds, as well as for 2378-TCDD, the compound of most specific concern.

Background

There are millions of combustion sources in the United States. Residential heating units burn oil, gas, coal and wood for heat. Larger commercial, institutional and utility boilers burn fossil fuels to generate heat and electricity. Many industrial processes burn fuels and other raw or waste materials to produce heat and/or recover products of marketable value. Other processes, such as incineration, use combustion to reduce the volume of unwanted waste products and to recover heat and other resources. Open fires, both accidental (e. g., structure and forest fires) and intentional (i.e., those set for forest management and agricultural burning), are other examples of combustion sources.

Assessment of CDD and CDF emissions from combustion sources has received limited study. Previous work included studies of emissions from hazardous waste incinerators, utility boilers and municipal waste combustors. Even for those

source categories that have been tested, there is considerable variation in the extent and quality of testing and in the test methods employed.

Study Design

It was impractical to test all of the combustion source categories under Tier 4. A study plan was developed that identified those source categories which were believed to have the greatest potential for emitting CDDs to the atmosphere. Selection and prioritization of source categories for testing were based upon a review of CDD related studies reported in the literature, and on engineering judgment.[2,3] Information from this review suggested that the following conditions were most important for CDD formation:

1. Presence of CDD in the materials being burned;
2. Presence of CDD precursors in the materials being burned (e.g., chlorinated phenols, chlorinated benzenes); and
3. Presence of chlorine, fuel and combustion conditions conducive to CDD formation, including:
   (a) Relatively low combustion temperature (500 - 800°C);
   (b) Short residence time of fuel in the combustion zone (< 1 to 2 seconds);
   (c) Lack of adequate oxygen (resulting in incomplete combustion);
   (d) Lack of adequate processing of fuels (e. g., burning of wet garbage); and
   (e) Lack of supplemental fuel to promote combustion efficiency.

Based on a relatively subjective determination of which combustion source categories were most closely associated with these factors, judgments were made as to the likely potential of various source categories to emit CDDs. Certain source categories judged to have a relatively low potential to emit CDDs were not given further consideration for testing. For example, process heaters and

gas turbines were believed to have a low potential because of their higher combustion efficiencies and use of fuels with low chlorine content (e.g., natural gas).[2]

Analysis of these combustion related conditions suggested that municipal waste combustors, sewage sludge incinerators and recovery boilers at kraft paper mills should be tested because these were judged to have potential for CDD emissions and because they were large source categories. Table 1 lists the source categories identified in the prioritization effort. A more thorough explanation of the selection and prioritization process is contained in the Tier 4 Project Plan, which was widely circulated for comment before implementation.[3] Some of the source categories in Table 1 were included primarily on the basis of reviewer's recommendations. A few source categories (wood stoves and mobile sources) were included since these sources were being tested for other purposes and the add on costs for CDD and CDF testing was small. Further adjustments were made to the initial list of sources to be tested as the study progressed.

Tier 4 sampling efforts focused on source categories that had not been widely tested. Although some municipal waste combustors were known to emit CDDs, no additional stack testing of this source category was performed.* Compared to most other source categories, a relatively large data base already existed. In addition, other air pollution control agencies, such as the New York Department of Environmental Conservation and Environment Canada were conducting or planning studies of municipal waste combustors. Selected stack emission data from municipal waste combustors are summarized later in this report. In addition, Tier 4 collected ash samples from municipal waste combustors.

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*Subsequent to this decision, Congress has directed EPA to provide a report specifically on municipal waste combustor emissions of CDDs under the requirements of Section 102 of the Hazardous and Solid Waste Act of 1984.

TABLE 1. COMBUSTION SOURCE CATEGORIES WHERE ASH AND STACK SAMPLES WERE COLLECTED

Source Categories Sampled	Sample Type	
	Ash	Stack[a]
Sewage Sludge Incinerator	X	X (3)
Kraft Paper Recovery Boiler	X	X (3)
Industrial Waste Incinerator	X	X (1)
Wire Reclamation Incinerator	X	X (1)
Secondary Copper Smelter	X	X (1)
Carbon Regeneration Furnace	X	X (1)
Drum and Barrel Furnace	X	X (1)
Wood Stove	X	X (1)
Wood Fired Boiler	X	X (1)
Mobile Source		X (2)
Charcoal Manufacturing Oven	X	
Utility Boiler	X	
Small Spreader Stoker Coal Fired Boiler	X	
Commercial Boiler	X	
Kiln Burning Hazardous Wastes	X	
Open Burning/Accidental Fires	X	
Sulfite Liquor Boiler	X	
Apartment House Incinerator	X	
Hazardous Waste Incinerator	X	
Hospital Incinerator	X	
Municipal Waste Combustor	X	
Charcoal Grill	X	

[a]Number in parentheses indicates the number of sources in the category which were stack tested under Tier 4.

Sample Collection

Two types of testing were considered for each of the source categories listed in Table 1, stack sampling and ash sampling.

1. Stack Sampling

Stack sampling provides the best quantitative measurement of CDD emissions; however, it is expensive (e. g., $50,000 to $100,000 per source, not including analytical costs). Where possible, stack gas samples were collected both before (inlet) and after (outlet) any pollution control device. Ash, feed and soil samples were also collected at sites that were stack tested.

Because of the high costs, only thirteen sources could be stack tested. Three kraft paper recovery boilers and three sewage sludge incinerators were tested because they appeared to have conditions particularly conducive to CDD formation. Only one source was tested in each of the other selected source categories. The focus of the testing program was primarily on sources believed to be indicative of average to worst case emissions situations.

2. Ash Sampling

Ash samples were primarily collected from air pollution control devices (flyash) or from the residues of combustion (bottom ash) to provide a general indication of the presense of CDDs. A secondary objective of the Tier 4 study was to examine possible relationships between ash and stack test results. If such a relationship could be determined, inexpensive ash samples could be used in lieu of expensive stack testing to identify source categories with high CDD and CDF emission rates. Use of ash data is currently limited because observed correlations between levels of CDDs in fly ash and CDD stack emissions are not sufficient for quantitative use.[2] Generally, ash samples were collected from three sources within each of the source categories listed in Table 1.

Site Selection

Selection of test sites for stack and ash sampling was based on a number of factors. EPA Regional Offices were asked to recommend candidate sources, based on criteria outlined in the Project Plan.[3] For stack sampling sites, a technical analysis was conducted to determine fuel composition and combustor operating parameters for a particular source category that would likely result in a "representative" to "worst case" emission situation. Candidate sources were then contacted, and pretest survey visits were made to identify plants with operations most closely resembling the hypothesized conditions and with acceptable stack sampling locations.

Once a site was selected for stack testing, a detailed test plan was prepared which described the physical layout of the source and specified the locations where samples would be collected. Each site specific test plan also identified the number and type of samples to be collected, the sampling methods to be used, and the quality assurance activities associated with that test site. These test plans were circulated for review. After the test was completed, a separate report for each site was prepared describing the actual testing performed and the test results.

Ash sampling sites were generally selected based upon recommendations from Regional, State and local environmental agencies. Ease of sampling and level of participation by the agencies were considered in those cases where several facilities appeared to be of equal interest. Ash samples were collected by State and local agencies and EPA contractors during the surveys of candidate sources for the stack sampling program, as part of actual stack sampling, and from selected additional facilities.

Sampling Procedure And Analyses

Consistent sample collection procedures were used at all sites. These procedures are described in three Tier 4 protocol documents. One document describes the ash sampling procedures; a second, the stack sampling procedures; and a third, the quality assurance measures and procedures.[4,5,6] The stack testing method used at Tier 4 sampling sites is, with minor modifications, the state of the art method proposed for use by a joint American Society of Mechanical Engineers (ASME) and EPA work group for municipal waste combustors. This procedure, which uses a modified EPA Method 5 sampling train, is described in detail in the stack test protocol document.[5]

EPA's "Troika" of three inhouse laboratories was responsible for the analysis, as well as for the preparation of the CDD and CDF analytical protocols and laboratory quality control procedures to be used with Tier 4 samples. Analytical methods are described in an addendum to a Troika procedures document.[7]

While the Troika was responsible for all CDD and CDF analyses, an EPA contractor, Radian Corporation, provided support for the analyses of other compounds. For example, samples of some of the fuels and other feed materials at each site were analyzed to determine the presence of possible precursors (e.g., chlorinated benzenes, biphenyls and phenols). In addition, continuous emissions data were collected for various stack gases (e. g., CO, CO_2, O_2) during each stack test. Procedures used in these analyses are described in a separate report.[6]

An independent quality assurance program was also conducted for the stack testing program, to ensure that test results were of acceptable quality. Another EPA contractor, Research Triangle Institute, conducted the quality assurance

program, which included both the auditing of three stack tests and the introduction of audit samples into the laboratories to evaluate their performance. The independent quality assurance program is described in a separate report.[8]

Results

Approximately 350 samples were collected, 20 - 25% of which were for internal quality assurance purposes. Thirteen sources were stack tested, and 72 sites were tested under the ash sampling program. Collected samples were sent to the appropriate analytical laboratory in accordance with established procedures.

1. Tier 4 Stack Test Results

Table 2 contains the CDD results of the 13 sites stack tested, while Table 3 presents the CDF results. Data presented in these tables represent concentrations of emissions measured in the stack gases. CDD/CDF stack concentrations have been normalized to an oxygen concentration of 3 percent. This removes the effect of dilution, and is a more appropriate means of comparing various combustion processes.

There is considerable variation in the concentrations among the sources tested under Tier 4. Each of the sources with valid data had detectable levels of CDDs and CDFs, although not all had detectable levels of 2378-TCDD. The reported 2378-TCDD, CDD and CDF concentrations from the secondary copper smelter are an order of magnitude or more larger than any other source tested under Tier 4, and as many as two to four orders of magnitude greater than concentrations from some of the sources. A number of sources have considerably lower concentrations than the secondary copper smelter, but considerably greater concentrations than a number of other sources. On the other hand, some sources (e. g., kraft paper recovery boilers) have very small concentrations of

TABLE 2. TIER 4 CDD STACK TESTING RESULTS
(ng/dscm @ 3% O_2)[a]

Source	2378-TCDD	Chlorinated Dibenzo-p-dioxin Homologues					
		Other Tetra-	Penta-	Hexa-	Hepta-	Octa-	Total[b] Tetra-Octa
Drum And Barrel Furnace	0.05	1.2	0.72	0.79	1.3	0.92	5.0
Industrial Carbon Regenerator	ND[c]	0.57	0.44	0.98	0.90	0.81	3.7
Industrial Waste Incinerator	4.5	77	100	150	230	61	630
Kraft Paper Mill Recovery Boiler							
Plant A	ND	ND	ND	0.06	0.18	0.49	0.73
Plant B	ND	ND	ND	0.10	0.26	0.83	1.2
Plant C	ND	0.13	0.15	0.39	0.88	1.4	2.9
Secondary Copper Smelter[d]	170	1400	2300	2200	5900	3700	16,000
Sewage Sludge Incinerator							
Plant A	0.05	11	0.18	0.51	2.5	5.3	20
Plant B	ND	0.40	ND	ND	0.22	0.98	1.6
Plant C	0.14	8.1	1.1	7.0	21	15	53
Wire Reclamation Incinerator[d]	0.07	1.2	2.2	14	130	290	440
Wood Fired Boiler (Salt Laden Wood)	0.28	47	48	49	39	11	200
Wood Stove	NR[e]	NR	NR	NR	NR	NR	NR

[a] ng/dscm @ 3% O_2 = nanograms per standard cubic meter of flue gas, normalized to 3 percent oxygen.
[b] Numbers across may not add up to totals, due to rounding.
[c] ND = Not detected, but generally at less than 0.1 ng/dscm @ 3% O_2.
[d] Estimated values. Stack sampling results for this site do not meet analysis quality assurance objectives, but do represent lower level estimates.
[e] NR = Not reported. Loss of data due to organic interferences.

TABLE 3. TIER 4 CDF STACK TESTING RESULTS
(ng/dscm @ 3% O_2)[a]

Source	2378-TCDF	Other Tetra-	Chlorinated Dibenzofuran Homologues				
			Penta-	Hexa-	Hepta-	Octa-	Total[b] Tetra-Octa
Drum And Barrel Furnace	0.90	14	6.2	3.0	2.0	0.55	27
Industrial Carbon Regenerator	ND[c]	1.2	0.37	0.59	0.61	0.54	3.3
Industrial Waste Incinerator	21	570	610	650	470	66	2400
Kraft Paper Mill Recovery Boiler							
Plant A	0.02	0.16	0.06	0.07	0.16	0.13	0.59
Plant B	0.01	0.13	ND	0.34	0.17	0.07	0.71
Plant C	0.01	0.46	0.46	0.59	0.50	0.09	2.1
Secondary Copper Smelter[d]	5100	18,000	19,000	6000	11,000	7200	65,000
Sewage Sludge Incinerator							
Plant A	NR[e]	33	10	0.10	0.5	0.10	44
Plant B	2.1	19	4.8	1.6	ND	0.07	28
Plant C	54	150	110	32	60	45	450
Wire Reclamation Incinerator[d]	0.40	29	22	65	230	230	580
Wood Fired Boiler (Salt Laden Wood)	1.8	37	23	13	6.5	0.92	83
Wood Stove	NR	NR	NR	NR	NR	NR	NR

[a] ng/dscm @ 3% O_2 = nanograms per standard cubic meter of flue gas, normalized to 3 percent oxygen.
[b] Numbers across may not add up to totals, due to rounding.
[c] ND = Not detected, but generally at less than 0.1 ng/dscm @ 3% O_2.
[d] Estimated values. Stack sampling results for this site do not meet analysis quality assurance objectives, but do represent lower level estimates.
[e] NR = Not reported. Loss of data due to organic interferences.

2378-TCDD, CDDs and CDFs. For most sources, the CDF concentrations appear to be related to those of CDDs (i. e., sources which emit high concentration of CDDs also emit high amounts of CDFs).

2. Quality Assurance

While the sampling and analysis methods used in this study were state-of-the-art, they are nevertheless evolutionary. During the course of the study, it was sometimes found that the analysis methods could not cope with high levels of interfering contamination from other pollutants which caused difficulty in achieving the desired validity and precision of results. Also, the stack sampling method is currently undergoing validation testing. Preliminary results indicate that recovery efficiencies from the sampling method may be low and variable, with possibly less than half of the CDDs and CDFs in the stack emissions being collected by the stack sampling method. Additional validation testing is currently underway.

The stack gas samples collected at the secondary copper smelter contained such high levels of CDDs and CDFs that the sensitivity of the analytical procedures and equipment employed was reduced. Therefore, the results for this source represent minimum levels, and actual values could have been considerably higher.*

At the wire reclamation incinerator, the levels of contamination from other organic compounds in the sample were so high, even after rigorous laboratory extraction and sample cleanup procedures, that only estimates of CDDs and CDFs are available. At the wood stove site, it could not be determined if CDD's and CDF's were present in any of the three stack test samples, due to

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*Subsequent to the Tier 4 test, the secondary copper smelter was retested by the source in conjunction with the State Agency. Results from this retest found CDD emissions to be one third of the Tier 4 results while CDF emissions were 70 percent of the Tier 4 values.[9]

similar organic contamination. No results were obtained from the mobile source exhaust samples because internal reference standards were not added to the samples prior to the extraction step in the analytical procedure. At a few other sites, relatively minor problems occurred with a limited number of samples but these did not affect the analysis or the overall integrity of the data.

3. Results Reported In The Literature

The scientific literature was reviewed to determine what combustion source stack test studies had been conducted that were similar in scope and measurement methodology to Tier 4.[10] CDD and CDF data for 17 sources in the United States and Canada are presented in Tables 4 and 5. These results have also been normalized to a 3 precent oxygen concentration.

Table 6 has been prepared to facilitate a comparison of these data with those obtained under the Tier 4 program. The sources in Table 6 are listed in descending order of 2378-TCDD concentrations. Eight source tests (seven coal fired boilers and one cofired boiler firing fuel and refuse) reported in the literature had "nondetectable" stack gas concentrations of CDDs and CDFs. Pre-1986 data for six municipal waste combustors are also provided.

4. Discussion Of Stack Test Results

Although it is useful to compare stack concentrations of CDDs and CDFs among sources, such a comparison does not provide information with respect either to the ground level concentrations that would result from these stack releases or to the relative differences in potential health effects of the various CDD and CDF homologues. This discussion addresses these points.

The determination of the ground level concentration includes the impact on dispersion of stack height, gas temperature, stack gas flowrate (i.e., the size of the source) and local meteorological conditions, in addition to CDD and CDF stack concentrations. These parameters were entered into the dispersion

TABLE 4. CDD EMISSIONS DATA FROM STUDIES SIMILAR TO TIER 4
(ng/dscm @ 3% O_2)[a]

Source	2378-TCDD	Chlorinated Dibenzo-p-dioxin Homologues					
		Other Tetra-	Penta-	Hexa-	Hepta-	Octa-	Total[b] Tetra-Octa
Hazardous Waste Incinerator	1.4	64	8.3	1.3	1.1	2.4	77
Hospital Incinerator	ND[c]	ND	74	65	79	110	330
Municipal Carbon Regenerator	ND	0.01	0.13	0.37	0.47	1.6	3.3
Municipal Waste Combustor							
Plant A	0.7	10	NR[d]	26	12	4.1	53
Plant B	26	700	1600	1700	1600	860	6400
Plant C	NR	2.1	1.7	3.4	25	14	46
Plant D	0.8	30	250	210	200	15	710
Plant E	16	640	1700	1200	520	210	4300
Plant F	NR	7	18	36	58	90	210
Cofired Boiler 80% Coal/20% Refuse	ND	ND	ND	ND	ND	ND	ND
Coal Fired Utility Boiler Seven Plants	NR	ND	ND	ND	ND	ND	ND

[a]ng/dscm @ 3% O_2 = nanograms per standard cubic meter of flue gas, normalized to 3 percent oxygen.
[b]Numbers across may not add up to totals, due to rounding.
[c]ND = Not detected, but generally less than 1 ng/dscm @ 3% O_2.
[d]NR = Not reported.

TABLE 5. CDF EMISSION DATA FROM STUDIES SIMILAR TO TIER 4
(ng/dscm @ 3% O_2)[a]

Source	2378-TCDF	Chlorinated Dibenzofuran Homologues					
		Other Tetra-	Penta-	Hexa-	Hepta-	Octa-	Total[b] Tetra-Octa
Hazardous Waste Incinerator	2.1	170	12	4.8	0.81	0.24	190
Hospital Incinerator	NR[c]	130	220	200	120	65	735
Municipal Carbon Regenerator	0.02	1.4	1.1	0.76	0.76	0.72	4.8
Municipal Waste Combustor							
Plant A	NR	150	NR	100	12	1.0	260
Plant B	310[d]	3300	4200	2200	1600	120	11,600
Plant C	NR	14	9.4	15	56	22	120
Plant D	4	65	60	13	3	ND[e]	150
Plant E	57	1400	2100	1400	400	41	5300
Plant F	NR	38	63	78	62	12	250
Cofired Boiler 80% Coal/20% Refuse	ND	ND	ND	ND	ND	ND	ND
Coal Fired Utility Boiler Seven Plants	NR	ND	ND	ND	ND	ND	ND

[a] ng/dscm @ 3% O_2 = nanograms per standard cubic meter of flue gas, normalized to 3 percent oxygen.
[b] Numbers across may not add up to totals, due to rounding.
[c] NR = Not reported.
[d] Only three of eleven tests conducted at this site report 2378-TCDF.
[e] ND = Not detected, generally at less than 1 ng/dscm @ 3% O_2.

TABLE 6. TIER 4* AND OTHER SOURCES LISTED IN RANK ORDER BY 2378-TCDD CONCENTRATION (ng/dscm @ 3% O_2)[a]

Source	2378-TCDD	Total CDDs	Total CDFs
*Secondary Copper Smelter[b]	170	16,000	65,000
Municipal Waste Combustor - Plant B	26	6,400	11,600
Municipal Waste Combustor - Plant E	16	4,300	5,300
*Industrial Waste Incinerator	4.5	630	2,400
Hazardous Waste Incinerator	1.4	77	190
Municipal Waste Combustor - Plant D	0.8	710	150
Municipal Waste Combustor - Plant A	0.7	53	260
*Wood Fired Boiler	0.28	200	83
*Sewage Sludge Incinerator - Plant C	0.14	53	450
*Wire Reclamation Incinerator[b]	0.07	440	580
*Sewage Sludge Incinerator - Plant A	0.05	20	44
*Drum And Barrel Furnace	0.05	5	27
Hospital Incinerator	ND[c]	330	735
Municipal Waste Combustor - Plant F	NR[d]	210	250
Municipal Waste Combustor - Plant C	NR	46	120
*Industrial Carbon Regenerator	ND	3.7	3.3
Municipal Carbon Regenerator	ND	3.3	4.8
*Kraft Paper Recovery Boiler - Plant C	ND	2.9	2.1
*Sewage Sludge Incinerator - Plant B	ND	1.6	28
*Kraft Paper Recovery Boiler - Plant B	ND	1.2	0.7
*Kraft Paper Recovery Boiler - Plant A	ND	0.7	0.6
Cofired Boiler (coal and municipal waste)	ND	ND	ND
Coal Fired Utility Boilers (7 Plants)	NR	ND	ND

[a] ng/dscm @ 3% O_2 = nanograms per standard cubic meter of flue gas, normalized to 3 percent oxygen.
[b] These values are estimated. The true values may be higher.
[c] ND = Not detected, generally at less than 1 ng/dscm @ 3% O_2.
[d] NR = Not reported.

component of the Human Exposure Model (HEM) to estimate the annual average ground level concentration in the vicinity of the source. An assumption made in the application of this model to the Tier 4 data is that the CDD and CDF emitted from the stack is a gas. The assumption of gaseous behavior is believed to be a reasonable one for these sources. While different calculated ambient air concentrations could result from consideration of particle deposition, it is believed that such effects would not be significant because (1) these sources are generally low level emitters and (2) the particle size is likely to be small enough that the effect of deposition on ambient air concentration will not be a significant factor.

EPA uses "2378-TCDD toxic equivalency factors" (TEF's) to compare the relative potency of one mixture of CDDs and CDFs with different mixtures of CDDs and CDFs.[11] The use of the TEF approach permits an estimation of the carcinogenicity of the mixture of CDD and CDF compounds relative to the carcinogenicity of 2378-TCDD. The TEFs for the various CDDs and CDFs used in this analysis are presented in Table 7.

The 2378-TCDD equivalents, calculated maximum ground level concentration and 2378-TCDD equivalent annual emissions for the Tier 4 sources, and for most of the sources from the literature, are presented in Table 8.* To place these results in some perspective, the cancer risk from inhalation exposure to a ground level concentration of 1 picogram per cubic meter of 2378-TCDD equivalence is estimated as 3.3 chances in 100,000, (i.e., 3.3×10^{-5}) assuming 70 years of continuous exposure.[12] The 2378-TCDD equivalent annual emissions is the

*Ground level concentration and annual emissions were not calculated for the eight sources with nondetectable CDD/CDF emissions. Neither the hospital incinerator nor the municipal waste combustor, Plant F, is included in Table 8.

TABLE 7. TOXIC EQUIVALENCY FACTORS USED IN ESTIMATING 2378-TCDD EQUIVALENTS

Compound(s)	Toxic Equivalency Factor
2378-TCDD	1.0
Other TCDDs*	0.01
Penta-CDDs	0.5
Hexa-CDDs	0.04
Hepta-CDDs	0.001
Octa-CDDs	0.000
2378-TCDF	0.1
Other TCDFs*	0.001
Penta-CDFs	0.1
Hexa-CDFs	0.01
Hepta-CDFs	0.001
Octa-CDFs	0.000

*In situations where 2378-TCDD or TCDF were not chemically analyzed in the sample, total TCDDs and TCDFs will have a relative potency factor of 1.0 and 0.1, respectively.

TABLE 8. TIER 4 AND OTHER SOURCES LISTED IN RANK ORDER BY 2378-TCDD EQUIVALENTS[a]

Source	2378-TCDD Equivalents (ng/dscm @ 3% O_2)[b]	2378-TCDD Equivalent Annual Average Maximum Ground Level Concentration[c] (Picograms/m^3)	2378-TCDD Equivalent Emissions (g/year)[d]
*Secondary Copper Smelter[e]	3900	1.5	800
Municipal Waste Combustor - Plant B	1400	9.1	500
Municipal Waste Combustor - Plant E	1300	1.5	140
Municipal Waste Combustor - Plant D	140	3.0×10^{-2}	95
*Industrial Waste Incinerator	130	1.2×10^{-2}	0.7
Municipal Waste Combustor - Plant A	56	0.30	80
*Wood Fired Boiler	29	6.1×10^{-2}	0.6
*Sewage Sludge Incinerator - Plant C	25	0.91	2
*Wire Reclamation Incinerator[e]	10	9.1×10^{-3}	1×10^{-2}
Hazardous Waste Incinerator	7.4	9.1×10^{-2}	2
Municipal Waste Combustor - Plant C	5.7	0.24	1×10^{-5}
*Sewage Sludge Incinerator - Plant A	1.3	3.0×10^{-3}	2×10^{-2}
*Drum And Barrel Furnace	1.2	6.1×10^{-5}	9×10^{-3}
*Sewage Sludge Incinerator - Plant B	0.52	1.2×10^{-3}	4×10^{-2}
*Industrial Carbon Regenerator	0.31	3.0×10^{-4}	2×10^{-2}
Municipal Carbon Regenerator	0.20	1.5×10^{-3}	4×10^{-5}
*Kraft Paper Recovery Boiler - Plant C	0.12	1.5×10^{-4}	0.3
*Kraft Paper Recovery Boiler - Plant A	0.01	3.0×10^{-5}	3.0×10^{-2}
*Kraft Paper Recovery Boiler - Plant B	<0.01	3.0×10^{-5}	3.0×10^{-2}

*Sources tested by Tier 4.

[a]Isomer-specific data are generally not available. Homologue data are considered to be composed of the most toxic isomers.

[b]ng/dscm @ 3% O_2 = Nanograms per standard cubic meter of flue gas, normalized to 3 percent oxygen.

[c]Ground level concentration calculation assumes compounds are present at analysis detection limits when reported as not detected (ND).

[d]Assumes 8160 operating hours per year.

[e]These values are estimated. The true values may be higher.

total burden to the environment from the stack for these sources. It differs from the maximum ground level concentration by being independent of atmospheric dispersion.

As with the stack concentration data presented in Tables 2 and 3, there is considerable variability among the various sources for all three of these parameters. In general, the sources with the highest stack concentrations of 2378-TCDD, CDDs and CDFs reported in Table 6 also had the highest ground level concentrations. One notable exception is the sewage sludge incinerator, Plant C. Stack concentrations at this plant are about two to three orders of magnitude less than those of the secondary copper smelter, yet the estimated ground level concentrations from the two sources differ by less than a factor of two. The sewage sludge incinerator has a relatively low stack with low temperature flue gas coupled with a high plant throughput, which leads to a relatively high ground level concentration impacting a small area very near the plant. On the other hand, the secondary copper smelter has a relatively tall stack with high temperature flue gas which results in a comparable ground level concentration, but at a significantly greater distance from the plant. The area impacted by this concentration is much greater.

In addition to estimating ground level concentrations, EPA has prepared a preliminary assessment of the potential cancer risks from inhalation exposure associated with emissions from these facilities. A detailed discussion of the risk assessment is not included in this report, however, due to the concerns raised by EPA's Science Advisory Board (SAB) during their review of the study. The SAB cautioned that risks were likely to be higher than estimated if other exposure pathways, in addition to inhalation, were considered (e. g., food chain) and if more sources had been tested. EPA agrees with these comments and

is currently developing a procedure to consider the risks associated with secondary pathways of exposure. Further testing of other sources may be considered as the Agency moves forward with its ongoing effort to decide whether CDDs or CDFs should be listed as a hazardous air pollutant.

The results of the Tier 4 stack test program, along with the preliminary risk assessments, have been provided to the appropriate State air pollution control agencies for their information and use.

5. Tier 4 Ash Sampling Results

Three different types of ash samples were collected: bottom ash, fly ash, and scrubber water effluent. Bottom ash is the residue left in the combustion chamber as a result of the combustion process. Fly ash is the material collected by air pollution control devices which would otherwise be released to the ambient air. Scrubber water effluent samples are samples obtained from wet scrubbers, an air pollution control device which uses water to filter both particulate and gaseous pollutants from the exhaust gas stream.

The results of the ash sample analysis for the 75 sites for Tier 4 are summarized in Tables 9 and 10. Table 9 presents data from the source categories with detectable values of 2378-TCDD equivalent while Table 10 is a listing of the source categories where 2378-TCDD equivalent was not detected in the ash. A total of 90 samples were analyzed from the 75 sites.

CDDs and CDFs were found in about one third of the bottom ash and fly ash samples and one half of the scrubber effluent samples. The highest concentrations were typically found in fly ash samples. Ash samples were collected from 21 different source categories. Twelve of the source categories had one or more ash samples with a detectable concentration.

It is presently difficult to interpret the significance of the ash data from an air pollution perspective. One of the objectives of the study was

TABLE 9. TIER 4 ASH SAMPLING RESULTS

Source Category/Source Sampled	Sample Type	2378-TCDD Equivalent (ppb)
Wire Reclamation Incinerator		
Source-C	Fly Ash	656[a]
Source-A	Fly Ash	87
Source-A	Bottom Ash	32
Source-A	Fly Ash	21
Source-A	Bottom Ash	4
Source-B	Fly Ash	0.3
Source-D	Fly Ash	ND[b]
Secondary Copper Smelter		
Source-B	Fly Ash	117[a]
Source-A	Fly Ash	13
Wood Fired Boiler		
Source-A	Fly Ash	158
Source-C	Fly Ash	135
Source-B	Fly Ash	51
Source-D	Scrubber Effluent	0.1
Source-A	Bottom Ash (2 Samples)	ND
Source-E	Fly Ash	ND
Source-F	Fly Ash	ND
Source-G	Fly Ash	ND
Source-H	Fly Ash	ND
Municipal Waste Combustor		
Source-C	Fly Ash	142
Source-D	Fly Ash	44
Source-B	Scrubber Effluent	4
Source-B	Scrubber Effluent	3
Source-B	Bottom Ash	0.3
Source-C	Scrubber Effluent	0.1
Hazardous Waste Incinerator		
Source-B	Scrubber Effluent	42.9
Source-A	Bottom Ash	ND
Source-C	Scrubber Effluent	ND
Carbon Regeneration Furnace		
Source-C	Fly Ash	18
Source-A	Fly Ash	0.1
Source-B	Scrubber Effluent	ND

TABLE 9 (CONTINUED). TIER 4 ASH SAMPLING RESULTS

Source Category/Source Sampled	Sample Type	2378-TCDD Equivalent (ppb)
Sewage Sludge Incinerator		
Source-C	Scrubber Effluent	8
Source-F	Scrubber Effluent	5
Source-B	Bottom Ash	0.1
Source-A	Bottom Ash	ND
Source-C	Bottom Ash	ND
Source-C	Scrubber Effluent	ND
Source-D	Scrubber Effluent	ND
Source-G	Bottom Ash	ND
Source-H	Bottom Ash	ND
Source-I	Bottom Ash	ND
Source-J	Scrubber Effluent	ND
Industrial Waste Incinerator		
Source-A	Bottom Ash	2
Commercial Boiler		
Source-B	Fly Ash	1
Source-A	Fly Ash	ND
Hospital Incinerator		
Source-D	Fly Ash	0.9
Source-B	Fly Ash	0.6
Source-A	Bottom Ash	0.4
Source-D	Bottom Ash	0.4
Source-C	Bottom Ash	ND
Drum and Barrel Furnace		
Source-B	Bottom Ash	0.5
Source-E	Bottom Ash	0.3
Source-C	Bottom Ash	0.2
Source-A	Bottom Ash	ND
Source-B	Bottom Ash	ND
Source-D	Bottom Ash	ND
Apartment House Incinerator		
Source-A	Bottom Ash	0.3
Source-B	Bottom Ash	0.1
Source-C	Bottom Ash	ND
Source-D	Bottom Ash	ND

[a]These values are estimated. The true values may be higher.
[b]ND = Not detected, generally less than 0.08 ppb.

TABLE 10. TIER 4 SOURCE CATEGORIES WITH BELOW DETECTION LIMIT ASH SAMPLE RESULTS[a]

Source Categories Sampled	Number of Samples		
	Fly Ash	Bottom Ash	Scrubber Effluent
Charcoal Grill	-	2	-
Charcoal Manufacturing Oven	2	1	-
Kiln Burning Hazardous Wastes	3	-	-
Kraft Paper Recovery Boiler	6	-	-
Open Burning/Accidental Fires	-	2	-
Small Spreader Stoker Coal Fired Boiler	3	1	-
Sulfite Liquor Boiler	-	-	4
Utility Boiler	3	-	-
Wood Stove	-	3	-

[a]Detection limit generally less than 0.08 ppb. Listed alphabetically.

to determine a correlation between fly ash and stack emission concentrations. While the presence of CDDs and CDFs in the fly ash appears to be a good indicator of the presence of CDDs and CDFs in the stack emissions, no quantitative relationship has yet been observed that could reliably predict the magnitude of CDD/CDF emissions in the stack gases.

A comparison of the data from sources with both fly ash and stack test samples is provided in Table 11. This table illustrates the apparent lack of correlation between the two types of samples. For example, the secondary copper smelter, which had significantly higher stack concentrations than any other source, has fly ash concentrations more than an order of magnitude lower than some other sources. Other sources with relatively low stack emissions had fairly high fly ash concentrations. At this time, ash data do not appear to be a reliable indicator of the relative magnitude of CDD/CDF emissions in the stack. Fly ash samples, on the other hand, are believed to be fairly reliable indicators of the presence of CDDs/CDFs in stack emissions.

The ash sampling results have been transmitted through EPA's Regional Offices to the appropriate State and local agencies for their consideration. Although of limited usefulness for air pollution control purposes, the data do provide a measure of the level of contamination in the ash that is disposed of as a solid waste.

Findings And Conclusions

This investigation included a review of information in the literature, as well as a special sampling program designed to collect data for combustion source categories believed to have the greatest potential to emit CDDs and CDFs. The findings from this investigation are presented below.

(a) CDDs and CDFs have been detected in the stack emissions from most, though not all, combustion source categories tested to date. All of the sources

TABLE 11. COMPARISON OF ASH AND STACK EMISSIONS

AT SOURCES WITH CONCURRENT MEASUREMENTS

Source	2378-TCDD Equivalents	
	Fly Ash (ppb)	Stack Emissions (ng/dscm @ 3% O_2)[a]
Wood Fired Boiler	158	29
Municipal Waste Combustor - Plant C	142	5.7
Secondary Copper Smelter	13	3900[b]
Industrial Carbon Regenerator	0.1	0.31
Kraft Paper Recovery Boiler C	ND	0.12
Kraft Paper Recovery Boiler A	ND	0.01

[a]ng/dscm @ 3% O_2 = Nanograms per standard cubic meter of flue gas, normalized to 3 percent oxygen.
[b]These values are estimated. The true values may be higher.

stack tested under Tier 4, and most of the combustion source categories tested by others reported in the literature, had detectable concentrations of CDD's and CDF's.

(b) There is considerable variability in the emission rates among source categories. For example, measured CDD emissions ranged more than four orders of magnitude from "nondetected" at seven coal fired power plants tested (detection limit at less than 1 ng/dscm) to approximately 16,000 ng/dscm of total CDDs at a secondary copper smelting facility. Most of the combustion source categories fell within an intermediate range, generally two to three orders of magnitude less than the concentrations at the secondary copper smelting facility.

(c) EPA has not yet determined the magnitude of the potential population risk from these sources. An effort is underway to consider risk from all routes of exposure (e. g., inhalation, ingestion, dermal contact) and for evaluating procedures for estimating nationwide impacts from these sources.

(d) The presence of CDD/CDF in the fly ash from a control device appears to be a good indicator of the likely presence of CDD/CDF in the stack emissions. However, at the present time, it does not appear that the ash samples can be used to reliably estimate the magnitude of CDD and CDF stack emissions from a source. Continued use of expensive stack test methods appears necessary.

Continuing Efforts

Although the Tier 4 study has been completed, the Agency plans a number of continuing efforts with respect to CDD emissions from combustion sources. These include:

(a) EPA has a project underway to respond to the requirements of Section 102 of the Hazardous And Solid Waste Act of 1984 concerning CDD emissions from municipal waste combustors. This effort is intended to identify design and operating guidelines to minimize CDD emissions.

(b) On July 7, 1987, EPA published in the Federal Register an Advance Notice of Intent to Propose Regulations on air emissions from new or modified municipal waste combustors under Section 111b of the Clean Air Act. EPA intends to regulate one or more designated pollutants thus invoking Section 111d of the Clean Air Act.

(c) EPA plans to continue its evaluation of CDD/CDF emissions from various sources. EPA has not yet determined whether CDD/CDF should be listed as a hazardous air pollutant under Section 112 or other wise regulated under other Sections of the Clean Air Act.

(d) EPA is continuing its efforts to standardize and refine stack sampling and analysis procedures to reflect improvements in the state of the art. The recommended ASTM stack test methodologies for municipal waste combustors are currently being validated by the Agency.

Additional Information

Including this report, a total of twenty-two reports have been published under this study. "National Dioxin Study Tier 4 - Combustion Sources" is common to the title of each report. Abbreviated titles together with the EPA Report Numbers are presented below.

TITLE	REPORT NUMBER
Project Plan	450/4-84-014a
Initial Literature Review	450/4-84-014b
Sampling Procedures	450/4-84-014c
Ash Sampling Program	450/4-84-014d
Quality Assurance Project Plan	450/4-84-014e

Quality Assurance Evaluation	450/4-84-014f
Project Summary Report	450/4-84-014g
Engineering Analysis Report	450/4-84-014h
Final Literature Review	450/4-84-014i
Test Report Site 1 SSI-A	450/4-84-014j
Test Report Site 2 ISW-A	450/4-84-014k
Test Report Site 3 SSI-B	450/4-84-014l
Test Report Site 4 BLB-A	450/4-84-014m
Test Report Site 5 BLB-B	450/4-84-014n
Test Report Site 6 WRI-A	450/4-84-014o
Test Report Site 7 WFB-A	450/4-84-014p
Test Report Site 8 BLB-C	450/4-84-014q
Test Report Site 9 CRF-A	450/4-84-014r
Test Report Site 10 MET-A	450/4-84-014s
Test Report Site 11 DBR-A	450/4-84-014t
Test Report Site 12 SSI-C	450/4-84-014u
Test Report Site 13 WS-A	450/4-84-014v

Readers seeking more detailed information should obtain the specific report(s) of interest. The Engineering Analysis Report (EPA-450/4-84-014h) is the primary detailed technical report resulting from the Tier 4 study.

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